

## Materials Science and Technology

### Nanomaterials

# Dynamics of Propagating Reaction Waves in Nanolaminate Foils

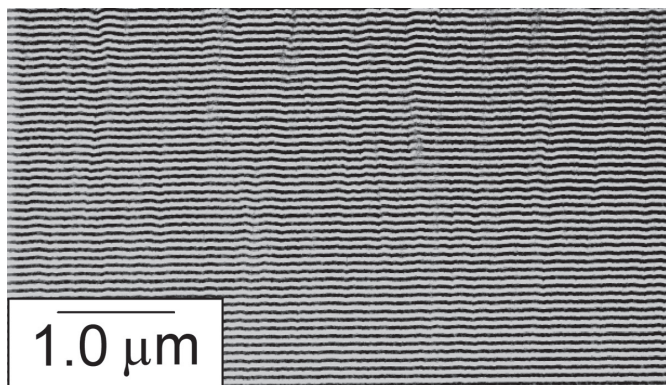


Figure 1: Cross-section, bright field transmission electron microscope image of an unreacted Ni/Ti nanolaminate foil. (Acknowledge: L. Brewer)

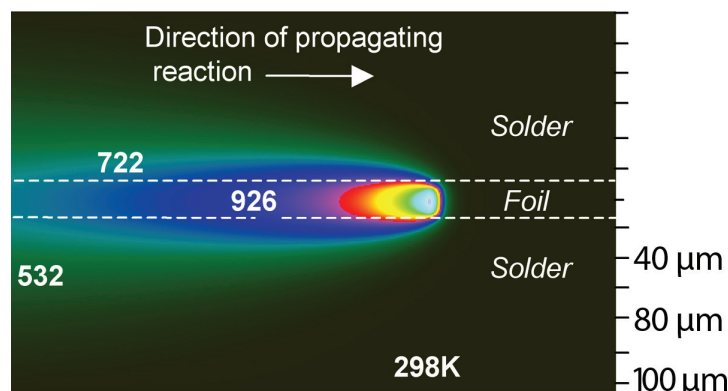


Figure 2: A simulated 'look' at a reaction front (shown in cross section) in a reactive Al/Pt nanolaminate constrained between two inert metals. Temperatures indicated are in degrees Kelvin.

*The extreme temperatures of reaction generated by laminate metal foils may find broad application in materials joining processes*

For more information:

**Technical Contact:**

David Adams  
505-844-8317  
dpadams@sandia.gov

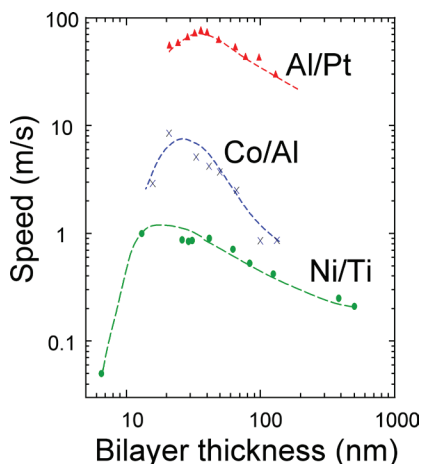
**Science Matters Contact:**

Alan Burns  
505-844-9642  
aburns@sandia.gov

**R**eactive nanolaminate foils have recently garnered interest for alternative joining processes that require localized soldering or direct attachment. As shown in Figure 1, they consist of alternating layers of metallic films, such as nickel (Ni) and titanium (Ti), that are fabricated by sputter deposition in a vacuum environment. Individual reactant layer thicknesses are precisely controlled, typically in the range of 5-500 nm; total foil thicknesses vary from 5-100  $\mu\text{m}$ , depending on the intended application. The finished foil is then applied to the surfaces that require joining. The chemical energy contained initially within a nanolaminate foil can be released as heat to melt and flow a neighboring filler material such as a gold-tin solder. By using only the heat evolved from a reactive nanolaminate for joining, there is virtually no thermal effect on neighboring temperature-sensitive devices or components, and minimal strain energy is developed in the assembly. For these reasons, exothermic nanolaminate joining processes have sparked interest in

the microelectronics industry for lid seal, die attach and bonding sputter deposition targets[1].

Reactive nanolaminates are of scientific interest because they exhibit extreme temperature fluctuations that can result in far-from-equilibrium crystal structures. A simulation of a propagating reaction and its cross-sectional temperature profile is shown in Figure 2. Furthermore, reactive nanolaminates grown by vapor deposition are an ideal class of materials for evaluating how reaction dynamics depend on reactant layer spacing and composition. Although it is well-established that the ignition thresholds[2] and reaction speeds of exothermic nanolaminates are directly affected by their nanometer-scale periodicity (see Figure 3), little is known about the dynamics of propagating reaction waves and how these events affect the final state of a foil. Understanding the reaction dynamics is essential to the aforementioned applications, because the manner in which heat is released influences the



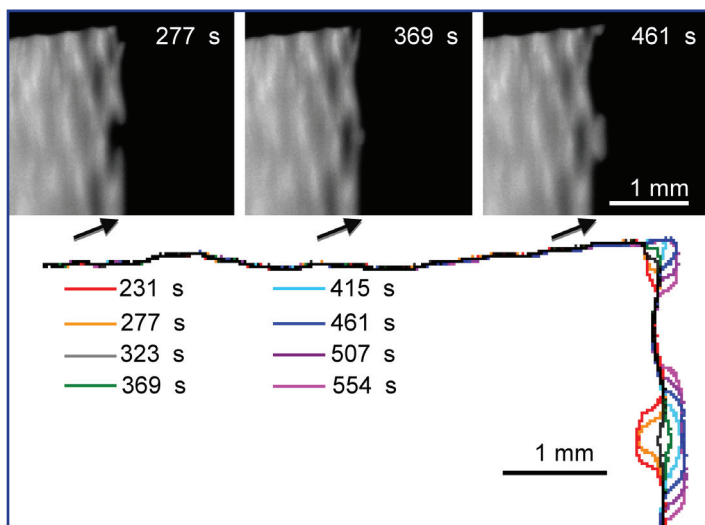
**Figure 3:** Plot of average propagation speed versus laminate periodicity for three exothermic material systems. Lines are guides to the eye.

microstructure, phase and surface morphology of the product foil and the structural integrity of a joined assembly.

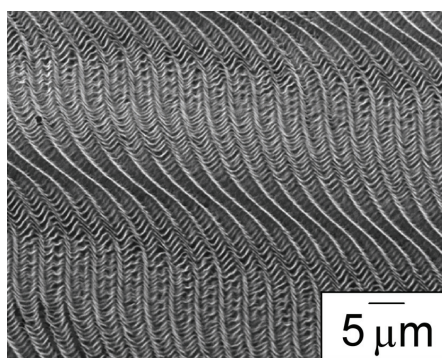
Sandia researchers and their collaborators[3] have investigated the dynamics of high-temperature, self-propagating reactions in metal-metal nanolaminates. Using both fast optical and *in situ* transmission electron microscopy, they have probed the evolving reactions and have determined how the shape of a reaction front depends on multilayer design, stored chemical energy, environment, and foil thickness. High exothermicity systems ( $\Delta H_o \approx -100$  kJ/mol) generally exhibit a microscopically-uniform reaction front with rapid average propagation speeds as high as 100 m/s. Low exothermicity systems ( $\Delta H_o \approx -50$  kJ/mol) exhibit unstable propagation modes characterized by a non-uniform reaction front morphology and reduced speeds. Unstable modes result from a decoupling of the atomic diffusion and thermal waves associated with a reaction front. In Figure 4, the shape of an unstable reaction front in a cobalt/aluminum (Co/Al) nanolaminate is demonstrated. Here, in plan view, the atomic diffusion wave lags the thermal wave, resulting in a propagating reaction mode that is characterized by transverse reaction bands originating at the boundaries of a specimen or at the intersections of colliding fronts [4]. This behavior is called a “spin-like reaction.”

The tendency to exhibit unstable spin-like behavior (versus stable modes) can have a direct effect on surface morphology and, in some cases, microstructure and phase formation. In Figure 5, scanning electron microscopy shows the surface morphology of a reacted Co/Al foil after it has cooled. In this case, a multi-periodic surface morphology developed as a result of spin-like propagating reaction waves with the wavelength of the final surface corresponding to the width of transverse propagating reaction bands.

Continued development of reactive nanolaminates for hermetic sealing and more broadly-applicable joining processes (potentially brazing) will benefit from an improved understanding of reaction dynamics, heat release rate and their effects on final foil properties including microstructure and morphology.



**Figure 4:** Time evolution of spin-like reaction waves in Co/Al. The three images show the surface of a reacting foil in plan view with reacted (hot) material appearing bright. Changes over time are observed at the reaction front (arrows). Spin-like behavior is evidenced by the propagation of reaction bands that move transverse to the net direction of the reaction wave (left-to-right). The color diagram depicts how colliding bands advance a reaction front a given distance ( $\sim 0.25$  mm), with continued reaction waves moving in opposite directions.



**Figure 5:** Scanning electron micrograph of a reacted Co/Al foil showing a rippled surface structure that is a direct result of propagating bands.

## References

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2. Y.N. Picard, J.P. McDonald, T.A. Friedmann, S.M. Yalisove and D.P. Adams: Nanosecond laser induced ignition thresholds and reaction velocities of energetic bimetallic nanolaminates, *Appl. Phys. Lett.* **93**, 104104 (2008).
3. A collaboration with T. Lagrange and G. Campbell (Lawrence Livermore National Laboratories).
4. J.P. McDonald, V. Carter Hodges, E.D. Jones Jr. and D.P. Adams: Direct observation of spin-like reaction fronts in planar energetic multilayer foils, *Appl. Phys. Lett.* **94**, 034102 (2009).